

Faceted anomalous scaling in the epitaxial growth of semiconductor films

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PACS 81.15.Aa – Theory and models of film growth

PACS 64.60.Ht – Dynamic critical phenomena

PACS 68.35.Ct – Interface structure and roughness

Abstract. - We apply the generic dynamical scaling theory (GDST) to the surfaces of CdTe polycrystalline films grown in glass substrates. The analysed data were obtained with a stylus profiler with an estimated resolution lateral resolution of $l_c = 0.3 \mu\text{m}$. Both real two-point correlation function and power spectrum analyses were done. We found that the GDST applied to the surface power spectra foresees faceted morphology in contrast with the self-affine surface indicated by the local roughness exponent found via the height-height correlation function. This inconsistency is explained in terms of convolution effects resulting from the finite size of the probe tip used to scan the surfaces. High resolution AFM images corroborates the predictions of GDST.

The kinetic roughening of interfaces is an outstanding topic of nonequilibrium Statistical Physics which has been intensively investigated in both theoretical [1–5] and experimental [6–10] frontlines. A generic dynamical scaling theory (GDST) for evolving interfaces includes both interface fluctuations and power spectra (structure factor) in the real and momentum spaces, respectively [2]. The analysis in the real space can be performed using the local height-height correlation function

$$G(l, t) = \langle [\overline{h(x+l, t)} - \overline{h(x, t)}]^2 \rangle, \quad (1)$$

where the overbar means averaging over the surface profile and $\langle \dots \rangle$ the averaging over distinct profiles. GDST gives that $G(l, t) = l^{2\alpha} \Phi(l/t^{1/z})$ where α and z are the roughness and dynamical exponents, respectively. The scaling function Φ behaves as

$$\Phi(x) \sim \begin{cases} x^{-2(\alpha-\alpha_{loc})} & \text{if } x \ll 1 \\ u^{-2\alpha} & \text{if } x \gg 1 \end{cases}. \quad (2)$$

The local roughness exponent α_{loc} determines the scaling locally. The power spectrum, defined as $S(k, t) = \langle \hat{h}(\mathbf{k}, t) \hat{h}(-\mathbf{k}, t) \rangle$ with \hat{h} being the Fourier transform of the height surface, scales as $S(k, t) = k^{-(2\alpha+d)} \Psi(kt^{1/z})$, where the scaling function Ψ is

$$\Psi(x) \sim \begin{cases} x^{2\alpha+d} & \text{if } x \ll 1 \\ x^{2(\alpha-\alpha_s)} & \text{if } x \gg 1 \end{cases}. \quad (3)$$

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In this scaling function, d is surface topological dimension, and α_s is the spectral roughness exponent.

This generic scaling ansatz has been observed in a large collection of models and experiments as can be looked up in [1, 6, 8] and references therein. This scaling ansatz implies a constraint between the exponents that depends specially on the spectral roughness exponent [1, 2, 11]. If $\alpha_s < 1$ the surface is self-affine with spectral and local roughness exponents being equal, $\alpha_{loc} = \alpha_s$. If $\alpha_s > 1$ the surface is locally smooth with $\alpha_{loc} = 1$. Each case is still classified into two subclasses. For $\alpha_s < 1$, if $\alpha_s = \alpha$ we have the regular Family-Vicsek (FV) scaling, otherwise the system has the intrinsically anomalous scaling. For $\alpha_s > 1$, we have the super-roughening scaling if $\alpha_s = \alpha$ and faceted growth scaling otherwise. The constraint between α_s and α_{loc} is a beautiful analytical result derived from the scaling ansatz [11] while the subclasses are allusive to their physical implications [1], with exception of the FV scaling which is due to the ansatz conceivers [12].

FV, super-rough, and intrinsic scalings have been reported in several experimental works [1, 6, 8] but the faceted one has only been achieved quite recently in the electrodisolution of pure polycrystalline iron [6]. The surfaces undergo a transition from intrinsic to faceted anomalous scalings as dissolution time increases. The surfaces in the faceted regime were highly anisotropic and, consequently, the scaling analyses were performed with one-dimensional profiles, in a direction orthogonal

to the anisotropy, in addition to the analyses of the two-dimensional surfaces. The faceted anomalous scaling was evident only for the one-dimensional case. This phenomenon was ascribed to the averages over all directions performed in $d = 2$, which underestimate the scaling exponents due to the contributions of unclear faceted morphology in some directions.

We investigate the anomalous roughening in semiconductor CdTe polycrystalline thick films grown on glass substrates. The real space scaling of this system was previously investigated [9, 13]. In the present work, we identify a crossover in the dynamical scaling exponents not noticed in Ref. [9] and perform the power spectrum scaling analysis. We find that the GDST applied to the surface power spectra foresees faceted growth whereas the correlation function analysis in points out for a self-affine surface. This inconsistency is explained in terms of the convolution effects resulting from the finite size of the probe tip used to scan the surfaces.

The CdTe films were deposited on glass substrates covered with a transparent conducting oxide (TCO), $\text{SnO}_2:\text{F}$, resulting a rough initial surface. The glass/TCO surface has a rms width of 16 nm that is much larger than 3 nm observed for the pure glass substrate. Although the presence of this layer introduces an undesirable large initial roughness, it is important from an application viewpoint because it is the front contact layer of solar cells produced with this material [14].

The samples used in the present work were produced by hot wall epitaxy at a growth rate of approximately 0.1 nm/s. Details of the growth system and sample preparation can be found elsewhere [9, 15]. It is important to mention that the growth of some samples was repeated using the molecular beam epitaxy technique and they exhibited the same behaviour. The scaling analysis was done in one-dimensional profiles with 300 μm of length and 4570 pixels acquired with a stylus profiler XP1-AMBIOS. Consequently, the topological (scan) dimension is $d = 1$ independently of the $2 + 1$ -dimensional surface. At least 20 profiles scanned at random directions were used in the averaging.

Despite of the good vertical sensibility (better than 1 nm) and step size scan (better than 100 nm) of the profiler the large tip radius ($\approx 1 \mu\text{m}$) limits the lateral resolution power due to convolution effects [16]. As a consequence, the data at short scales correspond to the convolution of the film surface and the probe tip geometries. A criterion accepted by the microscopy community establishes that a probe tip can resolve peaks separated up to 20% of its radius. Therefore, one can estimate a lateral resolution $l_c \approx 0.3 \mu\text{m}$ for this scan device. This lateral resolution, however, does not hinder the measurement of long wavelength fluctuations and the global roughness of the entire profile can be accurately determined. The short scale details of the surface morphology were visualized using an atomic force microscope (NTMDT-Ntegra Prima) operating in the semi-contact mode. These AFM images were

not used in the scaling analyses. The use of profiler scans allows to study large amplitude and wavelength fluctuations not achievable with a regular AFM. The growth time was varied from $t = 30$ to 660 min and the substrate temperatures $T = 150, 200, 250$, and 300°C were analysed. As will be shown, reliable scaling of the power spectrum were obtained for $T = 250$ and 300°C . The CdTe films detach from the glass substrates and crack during the cooling process for growth times longer than 660 min. This technical limitation hindered the growth of samples beyond this limit.

The exponents α_{loc} and α_s were obtained directly from the scaling behaviours of $G(l, t) \sim l^{2\alpha_{loc}}$ and $S(k, t) \sim k^{-(2\alpha_s+1)}$ at $t = 660$ min. The global interface width is defined as

$$W(t) = \left\langle [h(x, t) - \bar{h}(t)]^2 \right\rangle^{1/2} \sim t^\beta, \quad (4)$$

where β is the growth exponent. GDST states $\alpha_{loc} = \min(1, \alpha_s)$ and $\alpha = \beta z$ [2, 17]. Even though the local and spectral exponents are not independent, the local roughness exponent was measured for a comparison between the theoretical prediction and experimental observation.

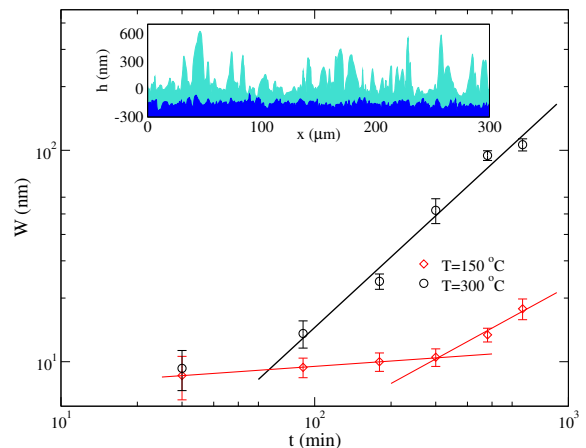


Fig. 1: Interface width evolution for two growth temperatures. Lines are power law regressions. Inset shows two surface profiles for growth times $t = 180$ min (dark) and $t = 660$ min (light) at $T = 300^\circ\text{C}$.

The surface width evolution, which is shown in Fig. 1, has a crossover from a regime of low to high growth exponents. This crossover was not perceived in the former scaling analysis of this CdTe/TCO/glass system, but it is also present as one can check in Ref. [9]. The crossover is probably on account of the initial surface roughness, which initially diminishes due to the occupancy of grooves and valleys. The temperature increase enhances the downward funnelling, reducing the crossover times and, consequently, the largest power-law interval was obtained for $T = 300^\circ\text{C}$. An additional evidence for our proposition is that this crossover was not observed in the CdTe deposition on pure glass substrates (a smaller initial roughness)

at the same experimental conditions [15]. Therefore, the growth exponents in Ref. [9] are under-estimated but the main conclusion of that work, stating a β exponent depending on temperature, still holds.

Whole scaling analyses were performed for the growth times after the crossovers. The β exponents are shown in Table 1. The large uncertainties for temperatures 150 and 200 °C reflect the short time intervals of power law growth. Taking into account the uncertainties, these exponents are nearly constant for temperatures 150, 200, and 250 °C. For $T = 300$ °C, the growth exponent $\beta > 1$ is a signature of an unstable growth that is unusually fast in the framework of kinetic roughening. However, self-affine (FV scaling) unstable growth was recently found in stochastic equations related to nonlocal interface dynamics [3,4].

Interestingly, the growth exponent obtained numerically for the unstable version of the stochastic Michelson-Sivashinsky (SMS) equation, $\beta_{sms} = 1.14$ [3], coincides with our estimate $\beta = 1.11(2)$. The number in parenthesis represents the uncertainty in last digit meaning $\beta = 1.11 \pm 0.02$. The mentioned unstable SMS equation, written in the momentum space with suitable rescaled parameters, reads as [3]

$$\partial_t \hat{h} = (k - k^2) \hat{h} + \frac{1}{2} \mathcal{F}(|\nabla h|^2) + \hat{\eta}, \quad (5)$$

where $\mathcal{F}(g) = \hat{g}$ is the Fourier transform of g and η a Gaussian noise. Apart from the term $k\hat{h}$, this equation is the well known Kardar-Parisi-Zhang equation [18] related to a non-conserved growth. The instability is due to the term $k\hat{h}$ that introduces the non-locality in the equation. However, as shown in the present work, the CdTe/Glass system is not self-affine ($\alpha_s \neq \alpha_{loc}$) and cannot be directly associated with the SMS equation.

Power spectra for $T = 300$ °C are shown in Fig. 2(a). The curves exhibit the usual plateaus for small and power law decays for large momenta. The power law yields $\alpha_s = 1.27(5)$ and, consequently, GDST states $\alpha_{loc} = 1$. Notice that momenta in the range $k < 2\pi/l_c$ were used in this analysis. Moreover, the curves shift upwards for large k as time increases, representing $\alpha > \alpha_s$ since $S(k, t) \sim k^{2\alpha_s+1} t^{2(\alpha-\alpha_s)/z}$ in the scaling ansatz given by Eq. (3). GDST thus predicts surfaces with anomalous scaling and facets. Even though the profiler provides coarse-grained scans that hinder to reliably resolve faceted morphologies, as can be seen in Fig. 3(a), a AFM image with a scan step size of 5.8 nm and probe radius of 10 nm and, therefore, a resolution about 100 times better than the profiler, clearly exhibits the faceted surface morphology predicted by GDST, as shown in Fig 3(b). That is a remarkable feat of the GDST which has foreseen the faceted morphology in coarse-grained data with a poor (if any) resolution of the facets. From a theoretical point of view, this result corroborates that GDST connects the scaling properties of long wavelength fluctuations with local morphological properties. In our case, the scaling properties of mesoscopic ($2\pi/l_c < k \ll 2\pi/L$) and macroscopic

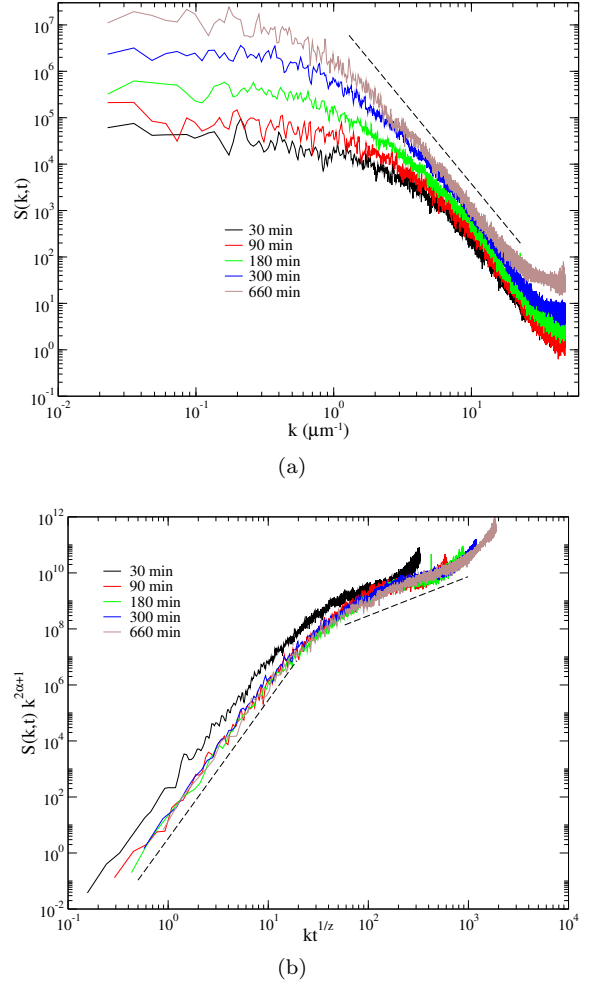


Fig. 2: (a) Power spectra of scanned profiles for $T = 300$ °C. Growth times increase from bottom to top. The dashed line has a slope -3.6 and represents the scaling $S \sim k^{-(2\alpha_s+1)}$. (b) Collapses of the power spectra using exponents $\alpha = 1.94$ and $z = 1.75$. Dashed lines represent the asymptotic behaviours of the scaling function $\Psi(x)$. The slopes are $2\alpha + 1 \approx 4.9$ and $2(\alpha - \alpha_s) \approx 1.3$.

($k \ll 2\pi/l_c$) scales are connected with the surface morphology in a microscopic scale $k > 2\pi/l_c$.

In Fig. 2(b) we probe the scaling ansatz for $\Psi(x)$, given by Eq. (3), by plotting $k^{2\alpha+1} S(k, t)$ versus $kt^{1/z}$, $\alpha = \beta z$, where $z = 1.75$ was obtained via gradient correlation function [explained in the next paragraph and shown in Fig. 4(a)] and $\beta = 1.11$ via global interface width (Figure 1). The excellent collapse obtained for $t \geq 90$ min provides an additional evidence that the data is described by anomalous scaling for faceted surfaces. Notice that the curve corresponding to $t = 30$ min, which was removed from the determination of the scaling exponents (Fig. 1), does not collapse. Finally, the asymptotic scaling forms of $\Psi(x)$ are also confirmed as indicated by the dashed lines.

The dynamic exponent z can be determined using the slope-slope correlation function $\Gamma(l, t) = \langle \nabla h(x + l) \nabla h(x) \rangle$. The correlation length ξ , which is assumed to

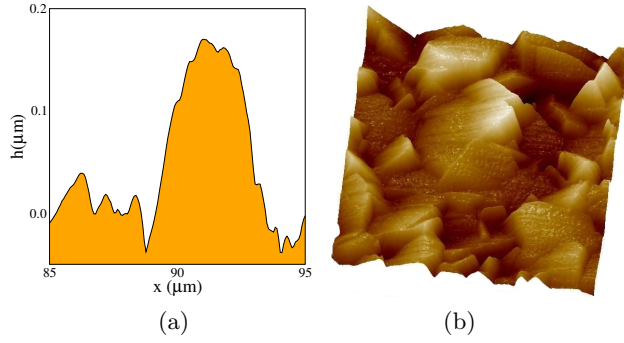


Fig. 3: Surface morphologies for CdTe after a growth time of 300 min at a temperature of 300 °C. (a) A 10 μm scan with using a profiler. (b) A 3 μm \times 3 μm AFM image illustrating the faceted CdTe surface.

scale as $\xi \sim t^{1/z}$ in GDST, can be defined as the first zero of the correlation function. The correlation functions and lengths are shown in Fig. 4(a). There is no clear power law regime with $\xi > l_c$ for $T = 150 - 250$ °C. For 300 °C, the slope provides $1/z = 0.57$.

Power spectrum analysis is itself sufficient to carry out the GDST, but correlation functions are widely used in the experimental investigations. Fig. 4(b) shows that the height-height correlation function, equation (1), also follows the usual qualitative behaviour of anomalous scaling, in which the curves are shifted upwardly as time evolves. However, a power law regression in the linear interval indicated turns out a self-affine surface with $\alpha_{loc} = 0.82(2)$, in disagreement with GDST. The scaling exponents for the other temperatures are shown in Table 1. The underestimated values of α_{loc} are reflecting the coarse-grained resolution of the scanning device since, in this scale, the analysed data are the convolution of surface and probe tip morphologies. The inset of Fig. 4(b) shows the correlation function collapses using the same exponents that collapsed the power spectra, as shown in Fig. 2(b). The collapses are so or more convincing than those obtained for power spectra.

The power spectra for $T = 250$ °C are shown in Fig. 5(a). The curve for $t = 600$ min also exhibits a power decay in a range within $k < 2\pi/l_c$ that yields $\alpha_s = 1.6(1)$. Again, $S(k, t)$ shifts upwardly as time increases implying $\alpha > \alpha_s$. Gradient-gradient correlation function does not provide a reliable estimate of the dynamical exponent z [Fig. 4(a)] and thus a criterion of best collapse for $t > 90$ min was used instead to obtain $z = 3.4(4)$ and $\alpha = 2.2(2)$. The collapse using these exponents are shown in Fig. 5(b). The power spectrum analyses for $T = 150$ and 200 °C did not provide reliable scaling regimes within the interval $k < 2\pi/l_c$. Even though the scaling for $T = 250$ °C is less precise than for $T = 300$ °C, it still points out a faceted anomalous scaling which was also confirmed in AFM images.

The power spectra for $T = 150$ and 200 °C do not exhibit scaling properties accurate enough to determine the

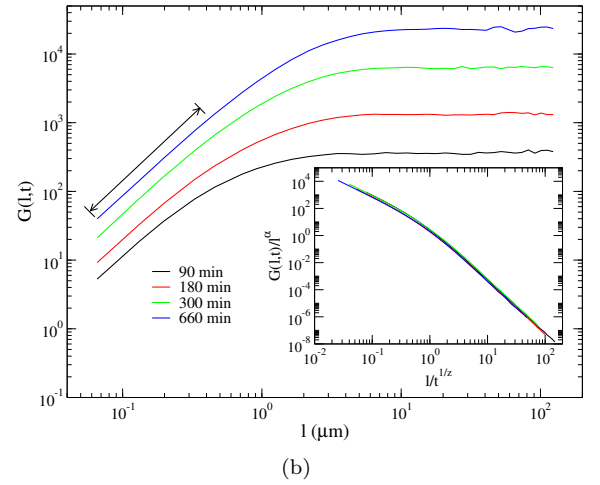
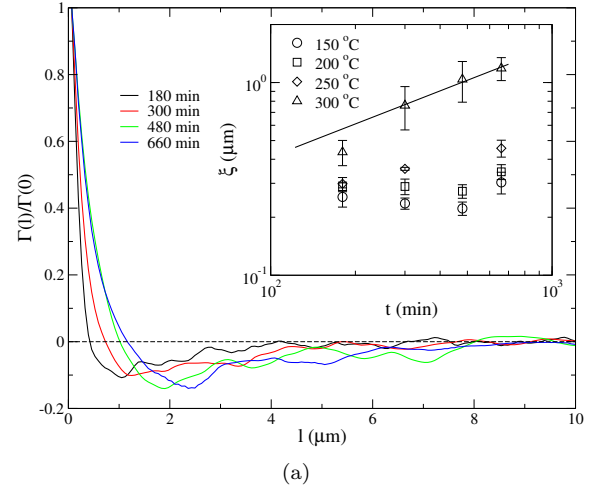


Fig. 4: (a) Slope-slope correlation functions for the CdTe surfaces. In the main plot, the correlation for distinct times and $T = 300$ °C are shown. Inset shows the correlation length against time. Straight line is a power law regression. (b) Height-height correlation functions for $T = 300$ °C. The interval of regression is indicated. Inset shows the collapse of the correlation functions using $\alpha = 1.94$ and $z = 1.75$

exponents but the qualitative behaviour of faceted anomalous scaling, $\alpha_s > 1$ and $\alpha > \alpha_s$, were also verified. AFM images again confirms the faceted morphologies for these temperatures. Indeed, the effect of temperature is to increase the size of faceted grains. For this reason, the scaling analysis with profiler data is more accurate for higher temperatures.

Now, we compare the the GDST in the momentum space with the results for real space of this CdTe-TCO-glass system reported in Ref. [13]. The scaling exponents α and z shown in Table 1 are in disagreement with those in Ref. [13], in which two sources of errors are present in the exponent determination. The first one is the underestimation of the growth exponent used to determine α via scaling relation $\alpha = \beta z$. The second one is the correlation length ξ that overestimated the dynamical ex-

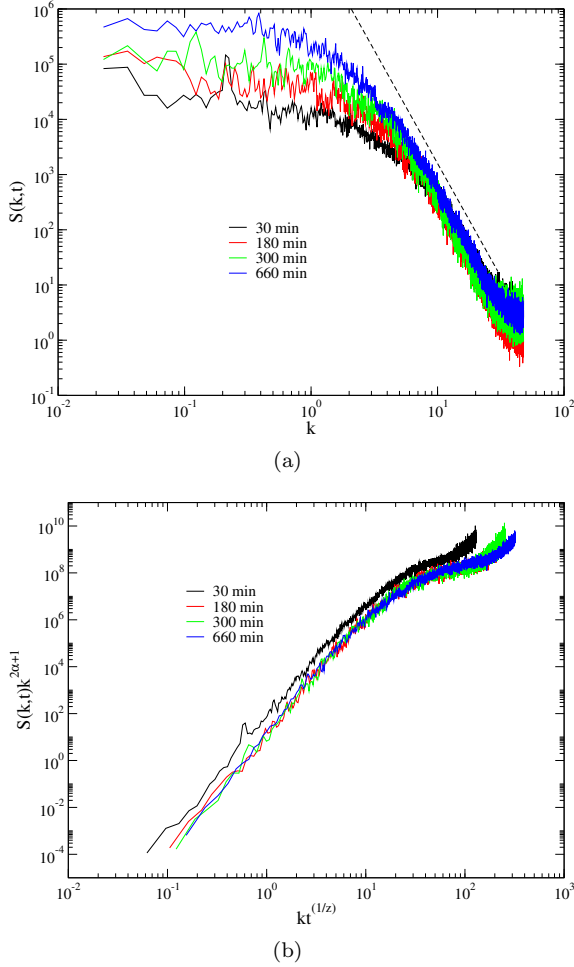


Fig. 5: (a) Power spectra of scanned profiles for $T = 250$ °C. Growth times increase from bottom to top. The dashed line has a slope -4.1 and represents the scaling $S \sim k^{-(2\alpha_s+1)}$. (b) Collapses of the power spectra using exponents $\alpha = 2.2$ and $z = 3.4$.

ponent z , as we clarify in this paragraph. The correlation length was defined in Ref. [13] as the characteristic length of the decay in a two-point correlation function $\Gamma(l, t) = \text{Pr}[|h(x+l) - h(x)| \leq m]$, where $m = 0.1|h_{\max} - h_{\min}|$ and $\text{Pr}[A]$ is the probability that the condition A is satisfied. The usual correlation function $\Gamma = \langle h(x+l)h(x) \rangle$ undergoes exactly the same effects. The correlation length was finally obtained by solving $\int_0^\xi \Gamma dl = 0.1 \int_0^\infty \Gamma dl$, where $\Gamma(l)$ is a two-exponential fit to the experimental data. However, if the tail corresponding to the long wavelength height fluctuations is or not left out of the regression, the exponents may change considerably. In Fig. 6, we compare the regressions of the correlation functions in two ranges: discarding (range 1) and including (range 2) the tail. The regression in the range 2, the same used in Ref. [13], misfits the data only for $l \lesssim 0.3 \mu\text{m}$ whereas the regression in the range 1 fits very well the small scales but deviates for $l \gtrsim 2 \mu\text{m}$, which was the reason for the choice of range 2 in Ref. [13]. The insertion to Fig. 6 shows the

Table 1: Scaling exponents for different growth temperatures. The numbers in parenthesis represent uncertainties in the last digit. The α_{loc} exponents were obtained using the height-height correlation function given by Eq. (1). The missing exponents are due to the absence of a reliable scaling regimes.

T (°C)	β	α_s	z	α_{loc}
150	0.59(9)	—	—	0.73(2)
200	0.56(9)	—	—	0.76(3)
250	0.65(2)	1.6(1)	3.4(4)	0.77(2)
300	1.11(2)	1.27(5)	1.75(5)	0.82(2)

correlation lengths against time and the respective power law regressions. Both cases yield quite satisfactory scaling laws, $\xi_1 \sim t^{0.58}$ and $\xi_2 \sim t^{0.27}$ corresponding to ranges 1 and 2, respectively. Assuming $\xi_i \sim t^{1/z_i}$, where $i = 1, 2$, we found $z_1 = 1.72$ and $z_2 = 3.69$. Notice that the regressions provided two characteristic lengths of the same magnitude ($1 < \xi_2/\xi_1 \lesssim 2$) but exhibiting very different scalings with time. Repeating this analysis for lower temperatures ($T = 150 - 250$ °C) we found $\xi_1 \lesssim 0.5 \mu\text{m}$ and no clear power law could be identified as observed for the gradient correlation function. Therefore, range 1 is the fit interval that yields dynamical exponents consistent with GDST.

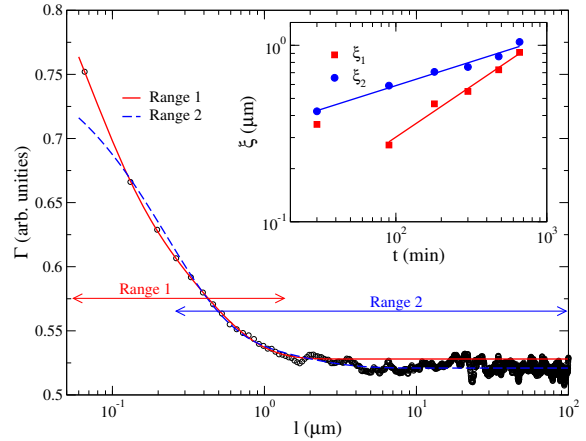


Fig. 6: Height-height correlation function of the CdTe surfaces. The growth time and temperature were 90 min and 300 °C. Circles represent the experimental data while solid lines the two-exponential regressions in the indicated intervals. Inset shows the correlation length obtained for the different regression intervals.

The conclusions of Ref. [13] are thus partially modified since we cannot apply GDST with those exponents to infer about which dynamical scaling regime the system belongs to. The exponent z presented in Ref. [13] was misleadingly associate to the dynamical exponent of the GDST. It would be better to refer to this exponent as a long wavelength coarsening exponent, which increases with temperature whereas the actual dynamical exponent

decreases. These results also stress out the difficulties usually observed in the fitting procedure of experimental data and the problems which can arise from a misinterpretation of the parameters obtained. In this scenario, GDST using momentum space appears as a robust method since local scaling properties can be obtained from mesoscopic and macroscopic scales.

In conclusion, we presented the scaling analysis of CdTe polycrystalline surfaces grown in glass substrates using a generic dynamical scaling theory (GDST) [2]. We investigated both height-height correlation functions and surface power spectra. The surfaces were scanned with a stylus profiler a resolution estimated as $l_c \approx 0.3 \mu\text{m}$.

Surface power spectra exhibit reliable scaling properties for $l > l_c$ at temperatures 250 and 300 °C, with a spectral roughness exponent α_s greater than 1 but different from the global roughness exponent α . The GDST foresees therefore faceted morphologies with local roughness exponent $\alpha_{loc} = 1$ that represents locally smooth surfaces. The height-height correlation function exhibits scaling only for $l \lesssim l_c$ and the local roughness exponents are in the interval 0.7-0.8, indicating self-affine surfaces as described in a previous study of this system [9]. However, high resolution AFM images confirm that the surfaces are faceted. The underestimated values of α_{loc} are reflecting the low resolution of the scanning device since, in this range, the analyzed profile is a convolution of the surface and probe tip.

It is worth to note that in a previous reports on anomalous scaling in faceted morphologies, investigated in the dissolution of polycrystalline iron [6], the faceted anomalous scaling was evident only for one-dimensional sections in a direction orthogonal to the anisotropy. Differently, our scaling analysis of polycrystalline CdTe consists in averaging over several randomly selected directions.

A renormalization group analysis of stochastic equations shows that the anomalous scaling cannot be present in local growth models [1]. Therefore, this theoretical result implies that disorder and/or nonlocal effects are responsible by the anomalous scaling in experimental systems. Indeed, the CdTe/TCO/Glass system undergoes both effects simultaneously. The amorphous glass substrate results in random growth orientations and, consequently, a polycrystalline film is obtained. Moreover, the glass substrate and CdTe films have distinct coefficients of thermal expansion implying strained film. The interplay between nonlocal strain and substrate disorder is a possible mechanism involved in the anomalous scaling.

Our work opens new paths to investigate anomalous scaling experimentally. A natural extension of this work, is to check the effect of the substrate on the scaling properties. Scaling analysis of AFM images may also complement the present investigation, in particular at lower temperatures where the scaling properties did not allow to determine the scaling accurately.

This work was supported by the Brazilian agencies CNPq, FAPEMIG and CAPES. SCF thanks the kind hospitality at the Departament de Física i Enginyeria Nuclear/UPC.

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